

REMARKS

Applicants are amending the previously considered claims in order to incorporate into claim 1 the subject matter of claim 6; to recite in claim 1 that R¹ and R² are each a methyl group; and to recite in claim 1 that X represents a hydrogen atom or an alkyl group. In light of amendments to claim 1, Applicants are cancelling claims 6 and 12 without prejudice or disclaimer, and are amending claims 8 and 14 to delete therefrom that R¹ and R² are each a methyl group. Applicants are amending claim 21 to correct a typographical error therein.

Moreover, Applicants are adding claim 22 to the application. Claim 22, dependent on claim 1, recites that X is a methyl group. Note, for example, Example 7 on page 13 of Applicants' specification, describing use of psudocumene as the aromatic compound represented by the general formula (1) (psudocumene being 1, 2, 4-trimethylbenzene).

Applicants respectfully request that the present amendments be entered, notwithstanding Finality of the Office Action mailed August 24, 2007. In this regard, noting previously considered claims 6 and 8, it is respectfully submitted that the present amendments to claim 1, as well as newly added claim 22, do not raise any new issues, including any issue of new matter; and, moreover, in light of incorporation of subject matter of previously considered claims into claim 1, it is respectfully submitted that the present amendments materially limit issues remaining in connection with the above-identified application. It is respectfully submitted that the present amendments are timely, in light of issues addressed by the Examiner in the Final Office Action mailed August 24, 2007, the present amendments being Applicants' first opportunity to amend their claims in light of contentions by the Examiner in the Office Action mailed August 24, 2007.

In view of the foregoing, it is respectfully submitted that Applicants have made the necessary showing under 37 C.F.R. §1.116(b)(3); and that, accordingly, entry of the present amendments is clearly proper.

Applicants respectfully submit that all of the claims presented for consideration by the Examiner patentably distinguish over the teachings of the prior art applied by the Examiner in rejecting claims in the Office Action mailed August 24, 2007, that is, the teachings of the U.S. Patents to Lien, et al, No. 2,868,854, to Olah, No. 3,766,286, and to Frey, No. 2,372,320, under the provisions of 35 U.S.C. §103.

It is respectfully submitted that these references as applied by the Examiner would have neither taught nor would have suggested such a process for producing an alkyl aromatic compound as represented by the general formula (2), in the present claims, including wherein an aromatic compound represented by the general formula (1), with R¹ and R² of the general formula (1) each being a methyl group, and X representing a hydrogen atom or an alkyl group, is alkylated with an olefin having 2-4 carbon atoms in the presence of a Broensted acid and without presence of a Lewis acid, with the resulting mixture being subsequently added with a Lewis acid and being subjected to isomerization in the copresence of the Broensted acid and the Lewis acid, with the Broensted acid being HF and the Lewis acid being BF₃.
Note claim 1.

Furthermore, it is respectfully submitted that these applied references would have neither taught nor would have suggested such a process for producing an alkyl aromatic compound represented by the general formula (2), as in the present claims, having features as discussed previously in connection with claim 1, and, in particular, wherein X is a hydrogen atom in the general formulas (1) and (2), as in claims 8 and

14, with R^0 being an isopropyl group in the general formula (2), as also in claims 8 and 14; or wherein X is a methyl group (see claim 22).

As can be appreciated, where R^1 and R^2 are a methyl group and X is a hydrogen atom, the starting material is m-xylene; and where R^1 and R^2 are each a methyl group and X represents a methyl group also, the starting material is a trimethylbenzene (e.g., 1, 2, 4-trimethylbenzene) in the alkylation reaction.

Furthermore, it is respectfully submitted that the teachings of these applied references would have neither disclosed nor would have suggested such process as in the present claims, having features as discussed previously in connection with claim 1, and, moreover, molar ratios of various components in the reaction isomerization as in claims 2, 4, 5, 10, 15 and 16; and/or temperature of the alkylation reaction or isomerization as in claims 3, 5, 9, 11 and 17-20; and/or wherein the olefin is selected from the group as set forth in claims 7 and 13; and/or wherein the alkylation and the isomerization are carried out in a same single reaction chamber (see claim 21).

The present invention relates to a process for alkylation of an aromatic (benzene) compound having two methyl substituents in the meta positions, by alkylating an aromatic (benzene) compound, having two methyl substituents in the meta positions, with a lower olefin such as ethylene or propylene. The produced alkyl aromatic (benzene) compound is useful as a raw material for various industries, such as in pharmaceuticals, agricultural chemicals, liquid crystals, functional pigments, solvents and monomers for engineering plastics.

In alkylating a substituted benzene compound having two methyl substituents on the benzene ring, the position (ortho, meta or para position) of the aromatic ring of the raw material to which an alkyl group is introduced is determined by the effect

of the functional groups on the reactant substituted benzene compound. When the desired compound is not in accord with the substitution orientation inherent to the functional group, however, a positional isomer of the desired compound is obtained as a product, and it is necessary to carry out a disproportionation reaction, called an isomerization reaction or a transalkylation reaction, in order to introduce the alkyl group to the desired position.

Various techniques have been used for the alkylation, as described on pages 1-3 of Applicants' specification. These alkylation reactions use, as a catalyst, a Lewis acid represented by aluminum chloride; and also well known is an alkylation reaction using a catalyst HF which is a Broensted acid. However, these previously proposed methods have various problems, including a problem in connection with separation and purification of HF used as the catalyst, and wherein the yield of the desired product is undesirably low, or conversion of the raw material is not satisfactory.

Against this background, Applicants provide a process for producing an alkyl benzene compound having substituents at the 3- and 5-positions, which is high in yield and in selectivity to the desired compound, and which permits easy and efficient recovery and recycling of the catalyst. As described on page 5 of Applicants' specification, the present inventors have found that the desired alkyl benzene compound having substituents at the 3- and 5-positions can be obtained in a stable manner with a high yield and a high selectivity, under mild and simple reaction conditions, by alkylating a benzene compound having two substituents at the meta positions with an olefin having 2-4 carbon atoms in the presence of a Broensted catalyst such as HF, followed by addition of a Lewis acid such as BF₃ and permitting intramolecular isomerization to proceed.

Applicants have found that a Lewis acid hinders the alkylation reaction, and, therefore, should not be used in the alkylation stage. In view thereof, Applicants provide a process achieving both effective alkylation and isomerization, by performing the alkylation in the presence of the Broensted acid and without the presence of the Lewis acid, with the Lewis acid subsequently being added and wherein the isomerization takes place in the copresence of the Broensted acid and the Lewis acid. Note the last full paragraph on page 7, and the paragraph bridging pages 7 and 8, of Applicants' specification.

As to advantages achieved according to the present invention, attention is respectfully directed to the Examples and Comparative Examples on pages 10-15 of Applicants' specification. This evidence in Applicants' specification must be considered in determining obviousness. See In re DeBlauwe, 222 USPQ 191 (CAFC 1984).

As can be seen in Comparative Examples 2-7 and as discussed in the last full paragraph on page 13 of Applicants' specification, it is seen that, when BF_3 is added in the alkylation reaction stage, good results including a high yield and high selectivity are not obtained, even when amounts of HF and propylene are changed. Note also the results discussed in connection with Comparative Example 8 on page 14 of Applicants' specification, wherein it is described that the product obtained had a yield of desired 3,5-dimethylcumene of 7%, and the selectivity was 13% and the yield of 2,4-dimethylcumene was 27%. In the Examples according to the present invention, much higher yield and selectivity were achieved.

It is respectfully submitted that the unexpectedly better results achieved according to the present invention, as seen in comparing the Examples and Comparative Examples in Applicants' specification, clearly support unobviousness of

the presently claimed invention, wherein the alkylation is performed in the presence of a Broensted acid and without the presence of a Lewis acid, with a Lewis acid subsequently being added and isomerization taking place in the copresence of the Broensted acid and the Lewis acid.

Lien, et al. discloses rearrangement of certain secondary alkylbenzenes, wherein an isomerization reaction is performed to shift position of substituent alkyl groups on the benzene ring without rearrangement of the alkyl group itself. The isomerization process comprises contacting, under substantially anhydrous conditions and in the substantial absence of reactive hydrocarbons, a di-secondary alkylbenzene selected from the class consisting of the ortho isomer, the para isomer and mixtures thereof with at least an effective amount of BF_3 , and an amount of liquid HF at least sufficient to form a distinct acid phase; the contacting is carried out at a temperature of not more than about -30°C ; and removing HF and BF_3 from the acid phase under conditions to substantially avoid rearrangement reactions and recovering from the reaction product mixture a di-secondary alkylbenzene fraction containing the meta isomer. Note the paragraph bridging columns 5 and 6 of this patent. See also column 1, lines 58-71; column 2, lines 35-40 and 48-52; and column 4, lines 57-64.

It is respectfully submitted that this reference does not disclose, nor would have suggested, such process as in the present claims, including, in addition to an isomerization step, the alkylation with an olefin of the compound of the general formula (1), with R^1 and R^2 thereof each being a methyl group, at meta positions, and especially wherein X is a hydrogen atom (see claims 8 and 14) or is a methyl group (see claim 22), in the presence of a Broensted acid and without the presence of a Lewis acid.

It is respectfully submitted that the additional teachings of Frey would not have rectified the deficiencies of Lien, et al., such that the presently claimed invention as a whole would have been obvious to one of ordinary skill in the art.

Frey discloses a process for the catalytic alkylation of hydrocarbons. This patent discloses admitting an alkylatable hydrocarbon to an alkylator, with concentrated or substantially anhydrous hydrofluoric acid being admitted. This patent discloses that for purposes of illustration benzene is chosen as the alkylatable hydrocarbon; but it is within the scope of the process described in the patent to alkylate also toluene or iso-paraffins such as isobutane or isopentane. All examples of this patent use benzene. See page 2, left-hand column, lines 13-22. Note also page 3, right-hand column, lines 22-31.

Initially, it is noted that Frey discloses an alkylation process to alkylate hydrocarbons, while Lien, et al. discloses an isomerization reaction "of certain secondary alkylbenzenes". It is respectfully submitted that one of ordinary skill in the art concerned with in Lien, et al., to rearrange certain secondary alkylbenzenes, would not have looked to the teachings of Frey. It is respectfully submitted that there would have been no motivation for combining the teachings of these references as applied by the Examiner, absent hindsight use of Applicants' disclosure, which of course is improper under 35 USC 103.

Furthermore, even assuming arguendo, that the teachings of these references were properly combinable, such combined teachings would have neither disclosed nor would have suggested the presently claimed method, including, inter alia, alkylating the aromatic compound represented by the general formula (1) (including wherein R¹ and R² are each a methyl group, at meta positions) with the olefin, and/or wherein the alkylation is performed in the presence of a Broensted acid and without

the presence of a Lewis acid, and advantages thereof, with subsequent addition of the Lewis acid whereby the isomerization is performed in the copresence of the Broensted acid and the Lewis acid. In this regard, it is again emphasized that Frey discloses production of ethyl benzene, each of the examples disclosing use of benzene as a reactant. It is respectfully submitted that the combined teachings of Lien, et al. and Frey would have neither taught nor would have suggested reaction (alkylation) of the aromatic compound represented by the general formula (1), wherein R^1 and R^2 thereof are each a methyl group, at meta positions, with an olefin having 2-4 carbon atoms, as in the present claims, and advantages thereof as discussed in the foregoing; and/or other features of the present invention as discussed in the foregoing including both processing steps being carried out in the same single reaction chamber, and advantages thereof.

Olah discloses a process for isomerizing paraffins and/or alkyl substituted aromatic hydrocarbons, the isomerizable hydrocarbons being selected from the group consisting of paraffins, alkyl substituted aromatic hydrocarbons, and mixtures thereof, the hydrocarbons being isomerized at low temperatures in the presence of a catalyst comprising (a) one or more Lewis acids of a specified formula, and (b) a strong Bronsted acid, preferably comprising a strong halogen-substituted acid such as fluorosulfuric acid, trifluoromethanesulfonic acid, trifluoroacetic acid or mixtures thereof. Note the paragraph bridging columns 1 and 2 of Olah. See also column 3, lines 50-52.

Frey has been previously discussed, and deficiencies thereof have been previously discussed.

Initially, and as with Lien, et al. and Frey, it is emphasized that Olah discloses an isomerization process, while Frey discloses an alkylation process. Specific

isomerizable hydrocarbons are described in connection with Olah. It is respectfully submitted that one of ordinary skill in the art concerned with in Olah, pertaining to an isomerization reaction, with specific hydrocarbons, would not have looked to the teachings of Frey; in particular, there would have been no reason from the combined teachings of these references, or in any other manner to one of ordinary skill in the art, to combine the teachings of these references. It is respectfully submitted that only through hindsight use of Applicants' disclosure, which is improper under 35 USC 103, would one have combined the teachings of Olah and Frey, as applied by the Examiner.

In any event, even assuming, arguendo, that the teachings of Olah and Frey were properly combinable, such combined teachings would have neither disclosed nor would have suggested the combination of alkylation and isomerization steps as in the present claims, of the aromatic compound presented by the general formula (1) as in claim 1, wherein R¹ and R² are each a methyl group, at meta positions as discussed previously, particularly the aromatic compound as in claims 8 and 14, wherein, additionally, X is H, or the aromatic compound as in claim 22, wherein X is a methyl group, with the alkylation being performed in the presence of a Broensted acid and without the presence of a Lewis acid, while the isomerization is carried out in the copresence of these acids, and advantages thereof; and/or other features of the present invention as recited in the present claims, including, inter alia, both processing steps are carried out in a same single reaction chamber, and advantages thereof.

It is acknowledges that Frey discloses at page 2, left-hand column, lines 20-22 that "it is within the scope of this invention to alkylate also toluene or isoparaffins such as isobutane or isopentane". Such disclosure in Frey, describing specific

materials to be alkylated “within the scope of this invention”, would have taught away from the presently claimed process, including alkylation of the aromatic compound represented by the general formula (1) in claim 1, wherein R¹ and R² are each a methyl group, at meta positions, as in all of the present claims, much less wherein X is a hydrogen atom (see claims 8 and 14) or wherein X is a methyl group (see claim 22).

The contention by the Examiner in the Office Action mailed August 24, 2007, that in view of the combined teachings of Lien and of Frey, or the combined teachings of Olah and Frey, it would have been obvious to have performed the alkylation in the presence of a Broensted acid and without the presence of a Lewis acid, with a Lewis acid subsequently being added with the isomerization being conducted in the copresence of the Broensted acid and the Lewis acid, as in the present claims, is respectfully traversed. It is respectfully submitted that in light of the combined teachings of either of Lien, et al, or Olah, with Frey, and for simplification of processing, one of ordinary skill in the art would have conducted the alkylation in the copresence of the Broensted acid and the Lewis acid. To the contrary, Applicants have found that, according to the present invention, unexpectedly better results are achieved by conducting the alkylation in the presence of Broensted acid and without the presence of a Lewis acid, with a Lewis acid subsequently being added and the isomerization taking place in the copresence of the Broensted acid and the Lewis acid. It is respectfully submitted that the unexpectedly better results achieved according to the present invention, as shown by the Examples and Comparative Examples in Applicants’ disclosure, provide a basis for patentability of the presently claimed subject matter.

In view of the foregoing comments and amendments, entry of the present amendments, and reconsideration and allowance of all claims then pending in the above-identified application, are respectfully requested.

To the extent necessary, Applicants petition for an extension of time under 37 CFR 1.136. Authorization is herein given to charge any shortage in the fees, including extension of time fees and excess claim fees, to Deposit Account No. 01-2135 (Case No. 396.44981X00), and please credit any excess fees to such deposit account.

Respectfully submitted,
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